

Doping dependence of the upper critical field in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ from specific heat

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Abstract. – The low-temperature specific heat of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) single crystals in magnetic field H up to 12 T has been examined over a wide range of doping ($0.063 \leq p \leq 0.238$). From this we have mapped the upper critical field H_{c2} of LSCO across the entire superconducting diagram. It is found that the H_{c2} shows a doping dependence similar to that of the critical temperature T_c . We have discussed the implications of the result and proposed that there may be an effective superconducting energy scale responsible for the H_{c2} behavior in the underdoped region.

Introduction. – Determining the fundamental parameters of the superconducting state in high- T_c cuprates is crucial to understanding the nature of the high-temperature superconductivity. The upper critical field H_{c2} is one such quantity which is directly correlated with the microscopic coherence length ξ . Convenient methods for determining H_{c2} mainly come from the resistive transport or magnetization measurements. For conventional low- T_c superconductors, their low or moderate H_{c2} enables one to draw the line of $H_{c2}(T)$ in the field-temperature phase diagram from the critical temperature T_c to $T \rightarrow 0$ K with the application of the magnetic field H to suppress the superconductivity. Hence the zero-temperature $H_{c2}(0)$ can be accurately accessed. Moreover, it is shown that the temperature dependence of $H_{c2}(T)$ in these systems can be well described by the Werthamer-Helfand-Hohenberg (WHH) theory [1]. According to this theory, the $H_{c2}(0)$ can be estimated by the slope dH_{c2}/dT in the vicinity of T_c . Therefore in many cases through investigating the behavior of $H_{c2}(T)$ near T_c one can also satisfactorily obtain the $H_{c2}(0)$ of the sample.

In contrast, for high- T_c cuprates the $H_{c2}(0)$ is inherently huge in parallel with their high T_c . In most cases the superconductivity could be removed only with very intense magnetic field H , which is not always accessible in experiments [2]. Thus most determination of $H_{c2}(0)$ in high- T_c cuprates relies on the extrapolation of the high-

temperature $H_{c2}(T)$ to $T \rightarrow 0$ K based on the WHH theory. However, recent considerable resistive transport measurements indicate that the mapped $H_{c2}(T)$ shows unusual low temperature upward curvature, which is inconsistent with the saturation predicted by the WHH theory [3]. This observation casts doubt on estimating $H_{c2}(0)$ using such extrapolation procedure.

Presumably the above difficulty is thought to arise from the strong superconducting fluctuations in high- T_c cuprates, especially for the underdoped region [4]. The recent Nernst effect and torque magnetometry experiments highlighted this proposal [5, 6]. It is found that the vortex Nernst signal or diamagnetic magnetization persists well above T_c , indicating the survival of superconducting correlations at high T though the coherent superconductivity has disappeared at T_c . By tracing the field scale at which the Nernst or diamagnetic signal vanishes, the H_{c2} has been defined which is higher than that determined in resistive transports and importantly does not become zero through the T_c .

In the H - T phase diagram, the $H_{c2}(T)$ line signifies the transition from the mixed state to the normal state. From the viewpoint of the specific heat (SH), associated with this transition is the increase of the electronic density of states (DOS) with increasing H and eventually the recovery of the normal state DOS at $H = H_{c2}$. Moreover, theory has suggested a robust connection between the DOS in the mixed state and that in the normal state [7].

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Thus this recognition actually implies that there is a well-defined way to evaluate the $H_{c2}(0)$ from SH [8]. In this letter we present an analysis of the low-temperature SH in a series of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) single crystals. The field-induced increase of the electronic SH in the mixed state has been quantitatively determined. Combination with the normal state electronic SH available in the literature, the $H_{c2}(0)$ has been extracted in a wide doping range across the whole superconducting phase diagram. It is found that the $H_{c2}(0)$ becomes larger as one moves from the underdoped region to the optimal doping point, and then falls with increasing doping in the overdoped region, forming a “dome” shape like the T_c . We have discussed the implications of this finding and proposed that there is an effective superconducting energy scale responsible for the $H_{c2}(0)$ behavior in the underdoped region.

Experiment. — We have performed the low-temperature SH measurement in LSCO throughout the entire superconducting phase diagram [9–11]. The samples used for the study were single crystals prepared by the traveling-solvent floating-zone method. The superconducting transition temperatures T_c , defined as the onset of the diamagnetic signal in the magnetic susceptibility, are summarized in table 1. The hole doping level of the sample p is simply regarded as the Sr concentration x , which ranges from 0.063 to 0.238. The low-temperature SH was carried out on an Oxford Maglab cryogenic system with a thermal relaxation technique [11, 12]. The data below 12 K were analyzed for magnetic field H up to 12 T with H parallel to the c axis of the sample.

Results and discussion. — From the raw data, we have reliably separated the electronic SH $C_{el} = \gamma T$ from other contributions such as the phonon SH and the possible Schottky anomaly [10, 11]. In magnetic field H , for a superconductor, there is an increase of the electronic SH in the mixed state, denoted as $\gamma(H)T$. To investigate the field dependence of the $\gamma(H)$ of the sample is a useful method to identify the symmetry of the superconducting gap. For a conventional s -wave superconductor, in the mixed state the electronic DOS comes mainly from the vortex core regions. Since the vortex number increases linearly as H increases, the C_{el} and hence the $\gamma(H)$ is proportional to H . For a d -wave superconductor, however, Volovik first pointed out that the electronic DOS is actually dominated by contributions from the outer regions of the vortex in a magnetic field [13]. While the vortex number increases linearly with H , the inter-vortex distance is inversely proportional to \sqrt{H} . Thus both effects result in the C_{el} of a d -wave superconductor showing a \sqrt{H} dependence. In experiments, the \sqrt{H} behavior of the C_{el} , that is, $\gamma(H) \propto \sqrt{H}$, has been observed in YBCO and LSCO [7, 11]. These have been taken as the bulk evidence for the d -wave symmetry of the superconducting gap in high- T_c cuprates. Figure 1 shows the H dependence of the $\gamma(H)$ for all doping samples. It can be seen the data are well described by $\gamma(H) = A\sqrt{H}$ (shown as the solid

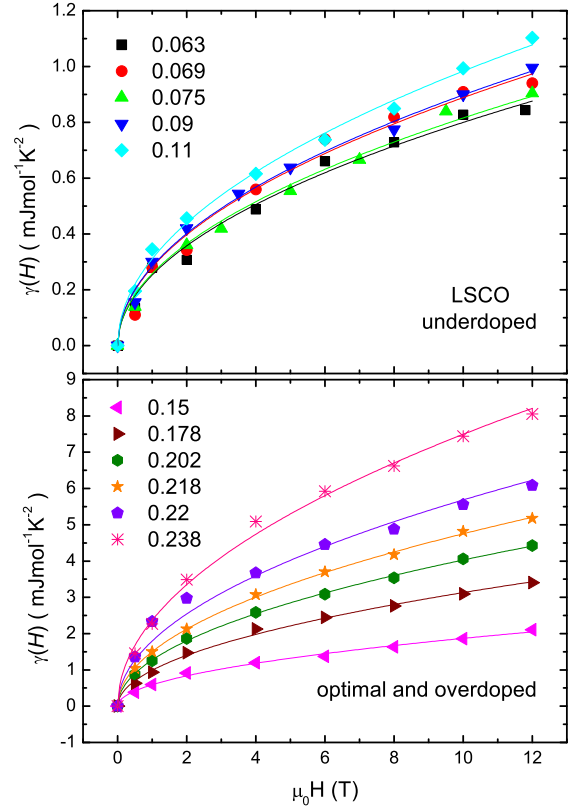


Fig. 1: Field dependence of the increase in the electronic SH, $\gamma(H)$, at $T \rightarrow 0$ K for all samples (symbols). Note that for $x = 0.238$ the $\gamma(H)$ has been corrected with the estimated superconducting volume fraction of the sample [10]. The solid curves are fits to the data with $\gamma(H) = A\sqrt{H}$.

curves) with A a doping-dependent constant. The numerical values of the prefactor A are listed in table 1. For completeness, the A for $x = 0.19$ reported by Nohara *et al.* is also included [14]. The above result suggests that the d -wave symmetry of the superconducting gap dominates the whole phase diagram of LSCO, which is consistent with the result of the recent phase-sensitive measurements [15]. Furthermore, we have shown that the field-induced SH is inversely proportional to the nodal gap slope v_Δ (and the gap maximum Δ_0), that is, $A \propto 1/v_\Delta \propto 1/\Delta_0$ [9]. From figure 1 and table 1 we can see the A essentially decreases with decreasing doping. Thus this suggests that the Δ_0 becomes larger towards underdoping [10]. Actually it has been shown that in the underdoped region, the Δ_0 quantitatively tracks the pseudogap in the normal state [9].

As the H keeps rising, the sample would inevitably undergo a phase transition into the normal state from the mixed state. In SH, this means the field-induced DOS increases and eventually the normal state electronic DOS is recovered. In other words, the $\gamma(H)$ rises and saturates to the normal state electronic SH coefficient γ_N at $H = H_{c2}$.

Table 1: Table I. Doping dependence of the H_{c2} for LSCO obtained from specific heat. The units of T_c , A , and H_{c2} are K, $\text{mJ mol}^{-1} \text{K}^{-2} \text{T}^{-0.5}$, and T respectively. The data for $x = 0.19$ are quoted from Ref. [14].

x	0.063	0.069	0.075	0.09	0.11	0.15	0.178	0.19	0.202	0.218	0.22	0.238
T_c	9.0	12.0	15.7	24.6	29.3	36.1	36.0	32.0	30.5	25.0	27.4	20.0
A	0.26	0.28	0.26	0.28	0.32	0.57	0.94	1.2	1.33	1.55	1.8	2.37
H_{c2}	16	17	39	53	75	82	56	45	45	44	36	17

Specifically, theory shows that for a d -wave superconductor the relation between the above two quantities can be written as

$$\frac{\gamma(H)}{\gamma_N} = \sqrt{\frac{8}{\pi}} a \sqrt{\frac{H}{H_{c2}}}, \quad (1)$$

where a is a constant depending only on the vortex lattice geometry ($=0.465$ for a triangular vortex lattice) [13, 16].

Combining eq. (1) with the experimental result $\gamma(H) = A\sqrt{H}$, we get $H_{c2} = 8a^2\gamma_N^2/\pi A^2$. This indicates that we may evaluate the $H_{c2}(0)$ from low-temperature SH provided we could also access the γ_N of the sample at $T \rightarrow 0$ K. Note that due to the large $H_{c2}(0)$ needed to suppress the superconductivity, the γ_N at $T \rightarrow 0$ K is hardly to be directly measured in SH for high- T_c cuprates. One alternative way is to investigate the superconducting transition of the sample and extrapolate the γ_N above T_c to $T = 0$ K based on the entropy conservation. In principle, we should do this performance in our own measurements. However, the uncertainty associated with the separation of the electronic SH from the phonon SH at elevated T makes our relaxation technique not well suitable for such a purpose. In this respect the community agrees that the differential calorimetry may give more accurate results [17]. Hence we instead look for the existing data in the literature. Actually, Matsuzaki *et al.* have recently estimated the γ_N at $T \rightarrow 0$ K of LSCO across the entire phase diagram in a systematic differential calorimetry study [18]. The doping dependence of the determined γ_N is reproduced in the inset of fig. 2. A roughly linear increase of the γ_N with increasing doping is established up to $x \simeq 0.2$. It is interesting to notice that the electronic DOS at the Fermi level in LSCO revealed by the angle-integrated photoemission spectroscopy (AIPES) follows essentially the same doping evolution [19].

For underdoped cuprates, we note that the above result implies there exists finite DOS in the ground state when superconductivity is suppressed at $T = 0$ K. Linked with the findings from the angle-resolved photoemission spectroscopy (ARPES), it is natural to speculate that this DOS resides on the Fermi arcs near $(\pi/2, \pi/2)$ nodal points for underdoped cuprates. ARPES has revealed that at T_c the Fermi surface is truncated forming arcs near the nodal regions in the pseudogap phase [20, 21]. It is further found that with increasing doping the length of the arc increases approximately linearly and at high doping level the arcs eventually connect with each other near $(\pi, 0)$ form-

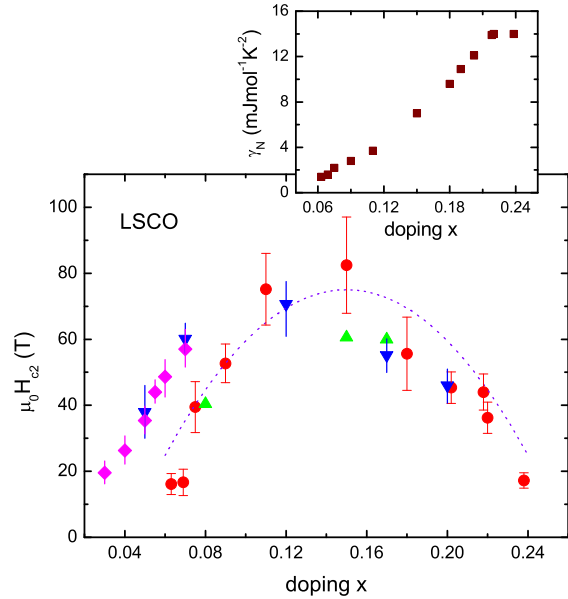


Fig. 2: Doping dependence of the upper critical field $H_{c2}(0)$ for LSCO obtained from SH (circles). The error bars are estimated from the uncertainties of the A values. $H_{c2}(0)$ forms a “dome” shape similar to the superconducting transition temperature T_c . For comparison, values of $H_{c2}(0)$ for LSCO determined from resistive transport (up triangles, Ref. [2]), Nernst effect (down triangles, Ref. [5]) and torque magnetometry (diamonds, Ref. [26]) are shown together. The dotted curve is a guide to the eye. Inset: Coefficient of the normal-state electronic SH γ_N at $T = 0$ K (Ref. [18]). They are cited for the evaluation of the $H_{c2}(0)$ shown in the main panel of the figure. See text for details.

ing a large Fermi surface [22]. Therefore, to reconcile with the SH it is not unreasonable to expect that for underdoped cuprates this Fermi arc state persists to $T = 0$ K as the ground state if the superconductivity was totally suppressed, which results in the finite DOS as shown in the SH [23]. Note that this notion has been supported by the independent nuclear magnetic resonance (NMR) study in $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+\delta}$ [24].

With the knowledge of the A and γ_N , we have derived the $H_{c2}(0)$ for LSCO according to the above expression, and plotted the result in the main panel of fig. 2. The

numerical values are also presented in table 1. The determination of $H_{c2}(0)$ in LSCO from specific heat allows us to see its general behavior in a wide doping regime. As shown in fig. 2, towards either underdoping or overdoping, the $H_{c2}(0)$ falls from its maximum value at optimal doping concomitant with the T_c , forming a similar “dome” shape as the T_c .

We have drawn the location of the $H_{c2}(0)$ in the H - p diagram for LSCO from SH. To gain more insight into the experimental result, let us compare our $H_{c2}(0)$ with that determined by other methods. Ando *et al.* have reported the c axis (interlayer) resistivity in intense magnetic field $H \parallel c$ for LSCO [2]. Choosing 90% of the normal-state resistivity as criterion, the H_{c2} was determined at low T and is plotted as the up triangles in fig. 2. We can see it shows good consistency with our result. Note that a similar “dome” shape of the $H_{c2}(0)$ in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$ (Bi2212) was also obtained from the high-field interlayer resistive transport [25]. In fig. 2, the down triangles and diamonds represent the H_{c2} for LSCO mapped in Nernst effect and torque magnetometry measurements, respectively [5, 26]. By extrapolating the high-field Nernst or diamagnetic signal to zero, the scale of H_{c2} has been determined. It is shown that, aside from very underdoped region, the H_{c2} obtained in Nernst effect and in SH agree reasonably with each other.

The quantitative agreement of the $H_{c2}(0)$ inferred from different methods supports the validity of our estimation of the $H_{c2}(0)$ from SH. Now we examine the implications of the experimental findings. From above we have shown $A \propto 1/\Delta_0$ and $H_{c2} \propto (\gamma_N/A)^2$, thus we obtain $H_{c2} \propto (\gamma_N\Delta_0)^2$. This indicates that the $H_{c2}(0)$ of the sample is governed by both the superconducting pairing strength and the DOS contributing to the superconducting condensation [23]. In the overdoped region, as shown in the inset of fig. 2, the γ_N increases slightly with increasing doping and finally becomes almost a constant. In spite of this, the $H_{c2}(0)$ is found to drop down monotonically as doping increases from the optimal doped point. This means that it is the reduction of the Δ_0 that dominates the behavior of $H_{c2}(0)$ in this region. In other words, the decrease of $H_{c2}(0)$ with overdoping should originate mainly from the reduction of the pairing strength.

Let us turn to the underdoped region, where the situation seems different. The SH measured in underdoped region has revealed that the Δ_0 continues growing in the underdoped region [9], which is also found by the thermal conductivity [27, 28]. In analogy with the overdoped side, one may expect the $H_{c2}(0)$ would keep rising with underdoping provided that the pairing strength is still the decisive factor to determine the $H_{c2}(0)$. Clearly this is at odds with the present experimental result which shows that the $H_{c2}(0)$ declines as doping reduces from the optimal doped point. Therefore, contrary to the overdoped region, this indicates that in the underdoped region it is the fall of the γ_N that overwhelms the rise of the Δ_0 and leads to the dropping down of the $H_{c2}(0)$ as the doping decreases.

In the above we have argued that in the underdoped region the γ_N in SH corresponds to the DOS on the Fermi arcs remaining in the pseudogap phase. Thus we can say for underdoped cuprates the $H_{c2}(0)$ represents the field scale necessary to recover the DOS on Fermi arcs from the superconducting state. Since the Fermi arcs shrink, that is, the DOS available to the superconducting condensation reduces towards underdoping, the $H_{c2}(0)$ naturally decreases concomitantly.

The above analysis actually indicates that in the underdoped region the coherent superconductivity below T_c may be triggered by the pairing of the carriers on nodal Fermi arcs. Since this pair process would be associated with the formation of an energy gap near the nodal region, it means that besides the pseudogap or the Δ_0 , there is an effective superconducting energy scale, denoted as Δ_{eff} , for underdoped cuprates. With decreasing doping, though the Δ_0 increases, the Δ_{eff} actually decreases since it represents the maximum gap on nodal Fermi arcs while the length of the Fermi arc reduces towards underdoping. For underdoped cuprates, the T_c or $H_{c2}(0)$ is just the temperature or field scale necessary to close this Δ_{eff} in a BCS-like fashion, respectively [29]. As one increases the T from below to $T = T_c$ or applies the field $H = H_{c2}(0)$, the Δ_{eff} closes and the carriers on nodal Fermi arcs are depaired, and therefore the coherent superconductivity is destroyed with the appearance of nodal Fermi arcs while the Δ_0 near $(\pi, 0)$ could remain unchanged. Note that this suggestion seems to be consistent with the very recent ARPES experiment which observed that a second energy gap opens at T_c and has a BCS-like temperature dependence in underdoped Bi2212 [30]. In contrast, for overdoped cuprates, however, the situation is much simpler. With $H = H_{c2}(0)$, the superconductivity disappears with the vanishing of the Δ_0 and the recovery of the DOS on the large Fermi surface.

In the very underdoped region, it is worth noting that the $H_{c2}(0)$ determined in Nernst effect and torque magnetometry seems to be larger than that determined in the present SH. This suggests that the $H_{c2}(0)$ probed by both methods may be different in this very region. We believe that it may originate from the strong superconducting fluctuations for underdoped cuprates [31]. In SH, since the H_{c2} marks the disappearance of the coherent superconductivity and the recovery of the normal state electronic DOS near nodal regions, it vanishes as the T_c is approached from the low temperature. However, due to sensitive to short-lived vortices, the Nernst effect or torque magnetometry may detect the fluctuating superconducting correlations above T_c and thus give a higher $H_{c2}(0)$ which remains a finite value at T_c . Note that a recent scanning tunnelling microscopy (STM) experiment in Bi2212 reported a nucleation of pairing gaps in nanoscale regions above T_c , which was considered as a microscopic basis for the above fluctuating superconducting response [32]. Under this circumstance, we view the $H_{c2}(0)$ defined in our SH as the field scale to destroy the phase coherence of

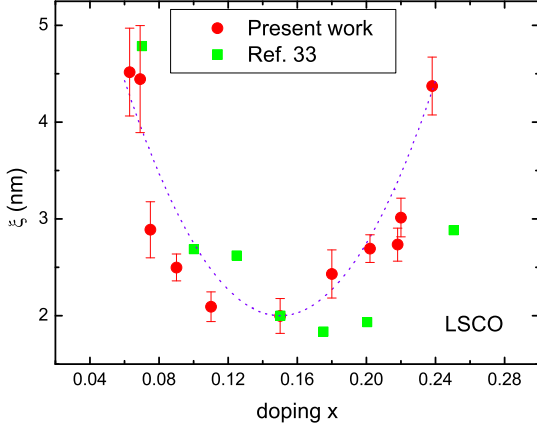


Fig. 3: Doping dependence of the coherence length ξ for LSCO derived from the H_{c2} with $H_{c2} = \Phi_0/(2\pi\xi^2)$ (circles). For comparison, the coherence length ξ obtained from magnetization measurements for LSCO (squares, Ref. [33]) is also plotted. The dotted curve is a guide to the eye. The results indicate a growing coherence length ξ from optimal doping point towards either underdoping or overdoping.

the superconductivity in the very underdoped region and thus have the suggestion that the pairing of the carriers on nodal Fermi arcs is crucial to and occurs simultaneously with the onset of the phase coherence. While at the same region the $H_{c2}(0)$ defined in Nernst effect or torque magnetometry may mark a higher field scale to destroy the phase-disordered condensate. Note that this is in particular indicated by the fact that, as shown in fig. 2, the $H_{c2}(0)$ probed in torque magnetometry varies continuously with x down to $x = 0.03$ while the coherent superconductivity only appears down to $x \approx 0.055$ [26]. On the other hand, it is interesting to note that for underdoped LSCO the $H_{c2}(0)$ probed in SH is actually roughly consistent in value with the field revealed in torque magnetometry which marks the melt of the vortex solid at $T \rightarrow 0$ K and falls to zero as $x \rightarrow 0.055$ [26]. In the end, We mention that the above speculation is also backed by the observation that in the overdoped region where fluctuations are most weak or absent the $H_{c2}(0)$ determined in different methods agree with each other.

Finally, from $H_{c2} = \Phi_0/(2\pi\xi^2)$, we can obtain the doping dependence of the coherence length ξ . Figure 3 shows the calculated ξ from the $H_{c2}(0)$ (circles). It is shown that from the optimal doping point the ξ grows towards underdoping or overdoping. Previously the doping dependence of ξ , regarded as the size of the vortex core, had been drawn from the systematic magnetization measurements in LSCO thin films [33], which is also plotted in fig. 3 (squares). It can be seen that both experiments show a reasonable consistency, which assures again the reliability of obtaining H_{c2} from low-temperature SH. More-

over, it should be noted that an increase of the ξ with underdoping from the optimal doping point has also been suggested by the fluctuation magneto-conductivity [34] and the reversible magnetization [35] measurements in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO). Furthermore, it could be instructive to compare ξ with the the Pippard coherence length $\xi_p = \alpha\hbar v_F/k_B T_c$, where v_F is the Fermi velocity and α a numerical constant of order unity [36]. As v_F is nearly doping independent [37], $\xi_p \propto 1/T_c$ and thus the variation of ξ_p with doping is qualitatively the same as that of ξ since both T_c and H_{c2} form a “dome” shape with doping. In terms of Δ_0 , on the other hand, ξ_p can be also expressed as $\xi_p = \hbar v_F/\beta\Delta_0$ with β another numerical constant. Note that there seems to be inconsistency between the above two expressions for ξ_p for underdoped region since T_c and Δ_0 have opposite doping dependence. Actually this seeming contradiction could be naturally resolved by substituting Δ_0 with Δ_{eff} in the expression of ξ_p , that is, $\xi_p = \hbar v_F/\beta\Delta_{\text{eff}}$ for underdoped cuprates. This further suggests that there may be an effective superconducting energy scale determining the coherent superconductivity in the underdoped region.

Conclusion. – In summary, we have analyzed the low-temperature SH in LSCO throughout the whole superconducting dome to evaluate the zero-temperature H_{c2} which is defined as the field scale necessary to remove the coherent superconductivity and recover the normal state electronic DOS. It is found that the doping dependence of $H_{c2}(0)$ essentially follows that of T_c . In the underdoped region, the decrease of $H_{c2}(0)$ concomitant with T_c suggests that the coherent pairing of the carriers on nodal Fermi arcs plays important role in establishing the high-temperature superconductivity.

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